

Collision dynamics between stretched states of spin-2 ^{87}Rb Bose–Einstein condensates

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Abstract We experimentally observed the time dependence of the spin populations of spin-2 ^{87}Rb Bose–Einstein condensates confined in an optical trap. The condensed atoms were initially populated in the stretched states $|F = 2, m_F = +2\rangle$ and $|F = 2, m_F = -2\rangle$ with several varieties of population imbalances. No spin-exchange collisions were observed in a weak magnetic field of 45 mG. The atom loss rate depended on the observed relative population of spin-states. We calculated the loss rate due to two-body inelastic collisions with the population imbalance using an experimentally estimated rate of $17.0(\pm 1.9) \times 10^{-14} \text{ cm}^3 \text{ s}^{-1}$ under the population balance. The calculations were in good agreement with the measurements. Our results show that the dependence of inelastic collisions on spin channels plays an important role in the time-evolution of spin populations.

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1 Introduction

Ultracold collisions of atoms play pivotal roles not only in degenerate gases but also in thermal atomic and molecular clouds [1]. In elastic collisions, the s -wave scattering length determines the properties of ultracold collisions [1, 2]. The s -wave scattering length can be controlled by the

magnetic Feshbach resonance [3] or the optically induced Feshbach resonance [4]. Inelastic collisions, especially two-body inelastic collisions [5], also play important roles in ultracold collisions for Bose–Einstein condensates (BEC) in the upper hyperfine state of the ground state of the alkali atoms [6–10], for sympathetic cooling [11–13], for buffer-gas cooling [14], for Penning ionization [15], etc. The inelastic collision rates depend on the magnetic field strength near a Feshbach resonance [16, 17]. For condensates in the upper hyperfine states, a high rate of inelastic collisions makes it difficult to observe relaxation effects that have been studied for condensates in the lower hyperfine state [18]. ^{87}Rb BEC is well suited to investigate the upper hyperfine state because the atom loss rate of ^{87}Rb is much smaller than that of other species [7].

There have been several experimental observations of the collision property of spin-2 spinor BEC [8–10]. Its magnetic ground state can be ferromagnetic, antiferromagnetic, or cyclic (spin-singlet trio) phases, whereas spin-1 BEC exhibits ferromagnetic or antiferromagnetic phases [19]. The phase in zero magnetic field is determined by the s -wave scattering length of the corresponding collision channels. Saito et al. [20, 21] proposed that the magnetic ground state can be determined from time-evolution measurement of the relative population of the spin-states in a weak magnetic field. In the cyclic phase, atoms initially populated in $|F = 2, m_F = +2\rangle$ and $|F = 2, m_F = -2\rangle$ states may produce $|F = 2, m_F = 0\rangle$ state atoms, while in the antiferromagnetic phase a $|2, \pm 2\rangle$ mixture is stable. The stability of the $|2, \pm 2\rangle$ mixture was experimentally observed by the Hamburg group [9]; however, the applied magnetic field of 340 mG was not weak enough to exclude quadratic Zeeman effects [20, 21].

In this paper, we investigate the temporal evolution of the spin-state populations of optically trapped spin-2 ^{87}Rb con-

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condensates in a weak magnetic field of 45 mG with various relative populations between $|2, +2\rangle$ and $|2, -2\rangle$ states. We estimate the atom loss rate caused by two-body inelastic collisions from population-balanced experiment and study the time-dependence of inelastic collisions on the relative populations in population unbalanced experiments. We also calculated the time-evolution of the number of atoms in the trap using a simple model of the density-dependent collisions.

2 Experimental setup

The experimental apparatus and procedure to create ^{87}Rb condensates are nearly the same as in our previous work except for some technical improvements such as the stability and accuracy of the computer control system [8]. A pre-cooled thermal cloud prepared by the first magneto-optical trap (MOT) was transferred to the second MOT by irradiating the first MOT with a weak near-resonant cw laser beam focused on the center of the atomic cloud. Over 10^9 atoms in the second MOT were pumped optically into the $|2, +2\rangle$ state and recaptured in an Ioffe–Pritchard (clover-leaf) type magnetic trap. BEC containing 10^6 atoms were created by evaporative cooling with frequency sweeping of an rf field for 15 s.

The BEC was loaded into a crossed far-off resonant optical trap (FORT) at a wavelength of 850 nm. One of the laser beams was irradiated along the long axis of the cigar-shaped BEC with a power of 7 mW and a $1/e^2$ radius at a focus of 24 μm . The other laser beam with a power of 11 mW and a beam waist of 90 μm was focused on and crossed the first beam. The potential depth of the crossed FORT was estimated to be about 1 μK . The power fluctuations of both lasers were less than 1%. The average trap frequency measured from the release energy of BEC [22] was found to be 82 Hz.

The condensed atoms in the $|2, +2\rangle$ state in the magnetic trap were transferred to the crossed FORT. The magnetic trap was then quickly turned off, and the quantization axis was nonadiabatically inverted. The condensed atoms were kept in the trap for 400 ms [8]. As a result, condensates containing 2.5×10^5 atoms in the $|2, -2\rangle$ state were prepared in the crossed FORT. To study the spinor dynamics, some of the condensed atoms in the $|2, -2\rangle$ state were transferred to the $|2, +2\rangle$ state by sweeping the rf field through resonance in an external magnetic field of 20.5 G [23]. In this magnetic field, the condensed atoms can be selectively transferred to the desired target states by quadratic Zeeman shifts. The relative populations of $|2, +2\rangle$ and $|2, -2\rangle$ states were controlled by the rf field strength and sweep speed. The observed fluctuation in the number of atoms was estimated to be 10% and resulted from variations in the initial number of BEC atoms and in the rf-transfer rate between spin states. After the state preparation, the 20.5-G field was

turned off, and an accurately controlled external magnetic field of 45 mG was immediately applied. The FORT was turned off at the end of the time-evolution period, and absorption imaging was applied after a free expansion time of 15 ms to measure the population distribution of each spin component by the Stern–Gerlach method [22]. A residual magnetic field of 10 mG was measured by observing the transfer rate for magnetic dipole transitions, and the residual gradient magnetic field was estimated to be 30 mG/cm [8].

3 Results and discussion

Figure 1(a) plots the time-dependence of the total number of condensed atoms initially in the $|2, +2\rangle$ and $|2, -2\rangle$ states with equal populations (closed triangles) and those occupying only the $|2, -2\rangle$ state (open squares). Each point was typically averaged over 4 samples. In these $|2, +2\rangle$ and $|2, -2\rangle$ measurements, the observed relative population of $m_F = +2$ ranged from 0.45 to 0.55. The total number of atoms in the $|2, +2\rangle$ and $|2, -2\rangle$ states in the trap rapidly decreased in comparison with that of atoms populated in the $|2, -2\rangle$ state alone because of hyperfine-changing inelastic collisions such as $|2, +2\rangle + |2, -2\rangle \rightarrow |1, m_F\rangle + |F, m'_F\rangle$ (where $F = 1, 2$). The observed condensate fraction was greater than 90%, and the temperature of the thermal cloud was below 100 nK. In the measurement, finite temperature effects in the condensates, as seen in [24], were not observed. The weak magnetic field strength of 45 mG satisfies the condition for suppressing the quadratic Zeeman effect [20].

The summation of relative population of $m_F = +2$ and -2 , that of $m_F = +1$ and -1 , and that of $m_F = 0$ are shown in Fig. 1(b). The summed relative population of $m_F = +1$ and -1 remained less than 0.05, that of $m_F = 0$ leveled off at approximately zero, and that of $m_F = +2$ and -2 remained stable above 0.94. This behavior suggests antiferromagnetic properties.

To study inelastic collisions between different spin states, we investigated the relation between the total number of atoms and the observed relative population of the $|2, +2\rangle$ and $|2, -2\rangle$ states for various trap times. The observed populations of other spin states were negligibly small. Figure 2 depicts the dependence of the total number of atoms on the observed relative population of $m_F = +2$ for trap times of (a) 0, (b) 50, (c) 120, and (d) 280 ms. The closed triangles were single-shot data for the observed relative populations. The open circles are averaged data for between 0.45 and 0.55 of the observed relative population. Under typical conditions, the initial population imbalance was $(50 \pm 20)\%$ as depicted in Fig. 2(a). As the trap time elapsed, the population imbalance grew. For example, at a trap time of 280 ms, it was $(50 \pm 45)\%$. The observed total number of atoms decreased more rapidly for smaller population imbalances.

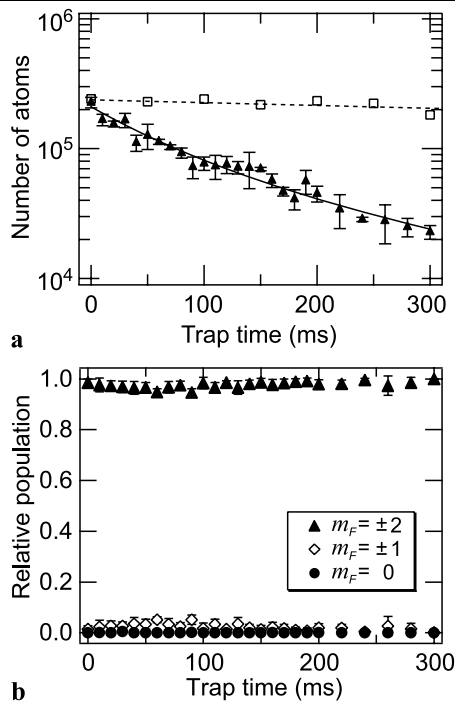


Fig. 1 Time-dependence of condensed atoms initially prepared in the $|2, +2\rangle$ and $|2, -2\rangle$ states. **(a)** Number of condensed atoms remaining in the trap as a function of trap time. *Closed triangles* represent the number of atoms initially populated in the $|2, +2\rangle$ and $|2, -2\rangle$ states, and *open squares* are that in $|2, -2\rangle$ state only. The experimental data are fitted by (1). **(b)** Time-evolution of relative populations of the summation of $|2, +2\rangle$ and $|2, -2\rangle$ atoms (*closed triangles*), $|2, +1\rangle$ and $|2, -1\rangle$ atoms (*open diamonds*), and $|2, 0\rangle$ (*closed circles*) atoms

This behavior can be understood qualitatively from the following property of the inelastic collisions. The collision rate for different spin states is higher than that for identical spin states. A major cause of two-body inelastic collisions between $|2, +2\rangle$ and $|2, -2\rangle$ atoms is hyperfine changing collisions in which a spin state of one or two atoms is changed to a lower hyperfine spin state [8]. Therefore, if only $|2, -2\rangle$ atoms are populated, no hyperfine changing collisions occur because of total spin conservation. In order to confirm this picture quantitatively, we calculated the population-dependent loss rate numerically.

The loss of atoms in a single-component BEC from the trap can be described by a differential equation as follows [5]:

$$\frac{dN}{dt} = -(K_1 + K_2\langle n \rangle + K_3\langle n^2 \rangle)N. \quad (1)$$

Here N is the total number of atoms, and K_1 is a rate coefficient for density-independent losses such as residual-gas scattering and photon scattering. K_2 and K_3 are the rate coefficients for two- and three-body inelastic collisions, respectively. In our experiment, the values of these coefficients were $K_1 = 0.25 \text{ s}^{-1}$ and $K_3 = 1.8 \times 10^{-29} \text{ cm}^6 \text{ s}^{-1}$ [8]. K_1 and K_3 can be ignored because their contribution to the atom

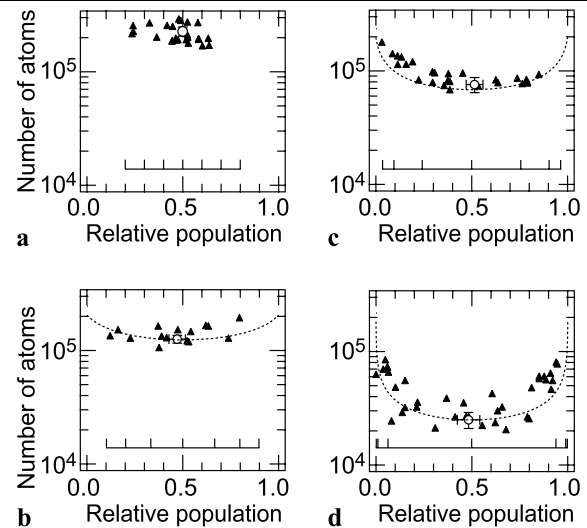


Fig. 2 Total number of atoms for various relative populations of $|2, +2\rangle$ (*closed triangles*) at trap times of **(a)** 0, **(b)** 50, **(c)** 120, and **(d)** 280 ms. *Open circles* represent the averaged data for observed relative populations from 0.45 and 0.55. The *dotted lines* are the calculation results for the population-dependent function of (3) with our experimental parameters. The *inset scales* show initial relative populations between 0.2 and 0.8 calibrated from (3)

loss is negligible in comparison with the K_2 term. Two-body inelastic collisions for two-component atomic gases change the atom density for $m_F = \pm j$ states, whereas the total atom density of single component gases is represented as $dn/dt = -K_2 n^2$ [25]. Thus, the loss rates due to two-body inelastic collisions are given by

$$\frac{dn_{\pm j}}{dt} = -K_{2(\pm j, \mp j)} n_{\mp j} n_{\pm j} - K_{2(\pm j, \pm j)} n_{\pm j}^2, \quad (2)$$

where $n_{\pm j}$ is the atom density of the BEC in the $\pm j$ state, $K_{2(\pm j, \mp j)}$ and $K_{2(\pm j, \pm j)}$ are the rate coefficients of two-body inelastic collision between $m_j = \pm j$ and $m_j = \mp j$ states and identical $m_j = \pm j$ state, respectively. When the Thomas–Fermi approximation is assumed for the atom densities of BECs, each component of the density can be represented by the relative population. Therefore, the rate equations for the number of atoms in $m_F = \pm j$ states are

$$\begin{aligned} \frac{dN_{\pm j}}{dt} = & -\gamma_2 N^{7/5} \rho_{-j}(t) \rho_{+j}(t) K_{2(+j, -j)} \\ & - \gamma_2 N^{7/5} \rho_{\pm j}^2(t) K_{2(\pm j, \pm j)}, \end{aligned} \quad (3)$$

$$\gamma_2 = \frac{15^{2/5}}{14\pi} \left(\frac{M\bar{\omega}}{\hbar\sqrt{a}} \right)^{6/5},$$

where $\rho_{\pm j}(t)$ is the relative population of the $m_F = \pm j$ states at the trap-evolution time t , M is the atomic mass, a is the scattering length, and $\bar{\omega}$ is the averaged trap frequency. γ_2 is derived from the relation between the number of atoms and the peak densities in the Thomas–Fermi ap-

proximation. It is assumed that any coherent spin dynamics can be ignored.

We focus on two-body collisions with $j = 2$, i.e., the collisions between $m_F = +2$ and -2 states. The inelastic collision rate in identical spin states, $K_{2(+2,+2)}$ and $K_{2(-2,-2)}$, can be ignored because $|2, +2\rangle$ and $|2, -2\rangle$ states are the stretched states. Thus, atoms inelastically collide with different spin state atoms selectively, while they do not inelastically collide with identical spin state atoms. As a result, we obtain a value of $K_{2(+2,-2)} = 17.0(\pm 1.9) \times 10^{-14} \text{ cm}^3 \text{ s}^{-1}$ in the experiment¹ and the fitted curve (solid curve) plotted in Fig. 1(a). The $K_{2(+2,-2)}$ value is somewhat larger than that for $m_F = 0$; $K_{2(0,0)} = 14(\pm 2) \times 10^{-14} \text{ cm}^3 \text{ s}^{-1}$ in a magnetic field ranging from 0.1 to 1.5 G [8]. These values are slightly larger than those of the Hamburg group ($K_{2(0,0)} = 10.2(\pm 1.3) \times 10^{-14} \text{ cm}^3 \text{ s}^{-1}$ and $K_{2(+2,-2)} = 13.2(\pm 1.8) \times 10^{-14} \text{ cm}^3 \text{ s}^{-1}$ at 340 mG)² [9]. The systematic uncertainty in estimating the experimental parameters such as the peak atom density and the trap frequency of the FORT may be the reason for this difference.

The dotted curves in Fig. 2 present the calculations of (3). They are in good agreement with the experimental results for various trap times. The calculations have no free parameters; we used the values of $N_0 = 2.1 \times 10^5$ and $K_{2(+2,-2)} = 17.0 \times 10^{-14} \text{ cm}^3 \text{ s}^{-1}$ determined from a population-balanced experiment. Figure 3(a) plots the time-evolution of the relative population for various initial population imbalances. Except for $\rho_{+2}(t) = 0.5$, the difference in the relative population increased with increasing the trap time. The relative population between 0.45 and 0.55 initially became 0.2 to 0.8 at 280 ms. The calculated imbalances were consistent with the experiment as shown in Fig. 2; the inset scales correspond to initial relative populations in the range of 0.2 to 0.8. The range increase results from dependence of two-body inelastic collision rates on spin channels; a pair of $m_F = +2$ and $m_F = -2$ atoms falls out of the trap together, whereas two identical spin state atoms do not cause a two-body inelastic collision. If the inelastic collision rate of the different spin state is the same as that of the identical spin state, the range of relative population would be held constant at all times. The thermal components were also negligible in our time-evolution experiment since the observed condensate fraction was larger than 90%.³ We cal-

¹In this condition, (3) can be simplified as $dN/dt = -0.5\gamma_2 K_{2(+2,-2)} N^{7/5}$, since the relative populations are equal, $\rho_{\pm 2}(t) = 0.5$. Note that the inelastic collision rate differs by a factor 2 from that in the identical spin state represented as $dN/dt = -\gamma_2 K_{2(j,j)} N^{7/5}$.

²The value is redefined by (3). The original value of $K_{2(+2,-2)}$ is $6.6(\pm 0.9) \times 10^{-14} \text{ cm}^3 \text{ s}^{-1}$ in [9].

³We calculated the number of atoms in two-body inelastic collisions in the presence of a thermal cloud, as described in [5]. The difference in the number of atoms for the calculations with and without thermal clouds was less than 5%.

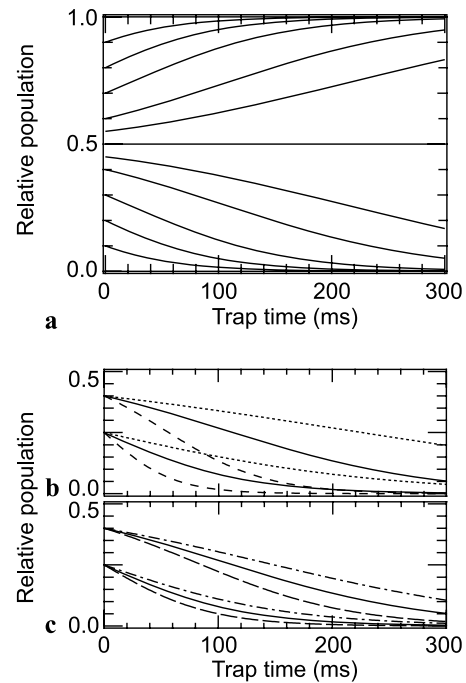


Fig. 3 Trap-time dependence of the relative population of $|2, +2\rangle$. (a) Time-evolution of the relative population for various initial population imbalances. (b) $K_{2(+2,-2)}$ dependence for initial populations of 0.25 and 0.4 at $N_0 = 2.1 \times 10^5$ and $K_{2(+2,-2)} = 17.0 \times 10^{-14} \text{ cm}^3 \text{ s}^{-1}$ (solid line), $8.5 \times 10^{-14} \text{ cm}^3 \text{ s}^{-1}$ (dotted line), and $34.0 \times 10^{-14} \text{ cm}^3 \text{ s}^{-1}$ (short-dashed line). (c) N_0 dependence for $K_{2(+2,-2)} = 17.0 \times 10^{-14} \text{ cm}^3 \text{ s}^{-1}$; $N_0 = 2.1 \times 10^5$ (solid line), 1.55×10^5 (dash-dotted line), and 4.2×10^5 (long dashed line)

culated the time dependence of the relative populations for various values of $K_{2(+2,-2)}$ and the N_0 dependence with initial population imbalances as shown in Figs. 3 (b) and (c), respectively. Comparing these figures, the dependence on $K_{2(+2,-2)}$ is more pronounced than the dependence on N_0 . Thus, fluctuations in N_0 smaller than 10% had negligible effect in estimating $K_{2(+2,-2)}$. In any case, the total spin of the condensate was conserved because inelastic collisions occurred selectively between spin-“+2” and spin-“-2” states.

As for the magnetic phase of the ground state, the cyclic phase has not yet been discovered, although its existence has been predicted for spin-2 BEC [19–21]. For the $F = 2$ system, the energy in an external magnetic field is characterized by the following function [19, 20]: $\epsilon_{\text{spin}} = c_1 \langle \mathbf{F} \rangle^2 + \frac{4}{3} c_2 |\langle s_- \rangle| - p \langle F_z \rangle - q \langle F_z^2 \rangle$, where \mathbf{F} , $\langle F_z \rangle$, and $\langle s_- \rangle$ denote the average spin, z component, and spin-singlet pair amplitude, and p and q correspond to the linear and quadratic Zeeman energies, respectively. Here c_1 and c_2 are parameters that characterize the spin-dependent mean-field energies and are defined by the s -wave scattering lengths $a_{\mathcal{F}}$ for binary collisions with total spin $\mathcal{F} = 0, 2, 4$ as $c_1 = (4\pi \hbar^2 / M)(a_4 - a_2) / 7$ and $c_2 = (4\pi \hbar^2 / M)(7a_0 - 10a_2 + 3a_4) / 7$. The cyclic phase occurs for $0 < c_2 < 20c_1$ and $|q| < c_2 n_{\text{total}} / 10$. Reference [20] assumed that $c_2 M / (4\pi \hbar)$

is at most of order of the Bohr radius and inelastic collision losses are spin-independent. To satisfy these conditions, a magnetic field of less than 100 mG is needed. The results of our measurements at 45 mG point to an antiferromagnetic ground state except when c_2 is less than the upper bound by the quadratic Zeeman energy q in the cyclic phase.⁴

It is primarily important to investigate inelastic collisions in order to understand the spin dynamics of BEC and to determine their magnetic phases. Using spin population dynamics, we need to characterize both the elastic and inelastic collisions in corresponding collision channels. In the cyclic phase, the $|2, +2\rangle\text{--}|2, -2\rangle$ collision produces $|2, 0\rangle$ atoms directly, while this collision does not produce $|2, 0\rangle$ atoms in the antiferromagnetic phase. Thus, the inelastic collision channels of $|2, +2\rangle\text{--}|2, 0\rangle$ and $|2, -2\rangle\text{--}|2, 0\rangle$ states should be considered. If the inelastic collision rate of $|2, \pm 2\rangle\text{--}|2, 0\rangle$ states is sufficiently high in comparison with other channels, it is difficult to distinguish the cyclic and antiferromagnetic phases because there are few newly produced $|2, 0\rangle$ atoms in the cyclic phase. In addition, a small number of $|2, +1\rangle$ and $|2, -1\rangle$ atoms exist in the trap due to imperfect transfer from $|2, -2\rangle$ to $|2, +2\rangle$. The channels $|2, \pm 1\rangle\text{--}|2, 0\rangle$ and $|2, \pm 1\rangle\text{--}|2, \mp 2\rangle$ should also be considered for determination of the ground state.

4 Conclusions

We observed the time-evolution of spin-2 ^{87}Rb condensates in $|2, +2\rangle$ and $|2, -2\rangle$ states. The $|2, 0\rangle$ state atoms do not appear within the trap time of 300 ms in a magnetic field of 45 mG. Our results for the spin population measurements in a weak magnetic field point to an antiferromagnetic ground state except when c_2 is less than the upper bound at 45 mG. We studied inelastic collisions of spin-2 ^{87}Rb condensates with population imbalances. The experimental results indicate that inelastic collision losses depend on the relative population. We calculated time-evolution of the relative population from simple two-body inelastic collisions that depend on the atom densities. The calculation results with no free parameters were in good agreement with the experiment for various trap times. A detailed study of the spin-stable dependence of inelastic collisions is a key issue for determination of the magnetic ground state of spin-2 ^{87}Rb BEC from spin-exchange dynamics.

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References

1. J. Weiner, V.S. Bagnato, S. Zilio, P.S. Julienne, *Rev. Mod. Phys.* **71**, 1 (1999)
2. N.N. Klausen, J.L. Bohn, C.H. Greene, *Phys. Rev. A* **64**, 053602 (2001)
3. S. Inouye, M.R. Andrews, J. Stenger, H.-J. Miesner, D.M. Stamper-Kurn, W. Ketterle, *Nature* **392**, 151 (1998)
4. M. Theis, G. Thalhammer, K. Winkler, M. Hellwig, G. Ruff, R. Grimm, J. Hecker Denschlag, *Phys. Rev. Lett.* **93**, 123001 (2004)
5. J. Söding, D. Guéry-Odelin, P. Desbiolles, F. Chevy, H. Inamori, J. Dalibard, *Appl. Phys. B* **69**, 257 (1999)
6. A. Robert, O. Sirjean, A. Browaeys, J. Poupard, S. Nowak, D. Biron, C.I. Westbrook, A. Aspect, *Science* **292**, 461 (2001)
7. A. Görlitz, T.L. Gustavson, A.E. Leanhardt, R. Löw, A.P. Chikkatur, S. Gupta, S. Inouye, D.E. Pritchard, W. Ketterle, *Phys. Rev. Lett.* **90**, 090401 (2003)
8. T. Kuwamoto, K. Araki, T. Eno, T. Hirano, *Phys. Rev. A* **69**, 063604 (2004)
9. H. Schmaljohann, M. Erhard, J. Kronjäger, M. Kottke, S. van Staa, L. Cacciapuoti, J.J. Arlt, K. Bongs, K. Sengstock, *Phys. Rev. Lett.* **92**, 040402 (2004)
10. A. Widera, F. Gerbier, S. Fölling, T. Gericke, O. Mandel, I. Bloch, *New J. Phys.* **8**, 152 (2006)
11. C.J. Myatt, E.A. Burt, R.W. Ghrist, E.A. Cornell, C.E. Wieman, *Phys. Rev. Lett.* **78**, 586 (1997)
12. G. Modugno, G. Ferrari, G. Roati, R.J. Brecha, A. Simoni, M. Inguscio, *Science* **294**, 1320 (2001)
13. D.J. Larson, J.C. Bergquist, J.J. Bollinger, W.M. Itano, D.J. Wineland, *Phys. Rev. Lett.* **57**, 70 (1986)
14. J.D. Weinstein, R. de Carvalho, T. Guillet, B. Friedrich, J.M. Doyle, *Nature* **395**, 148 (1998)
15. H. Katori, F. Shimizu, *Phys. Rev. Lett.* **73**, 2555 (1994)
16. J. Stenger, S. Inouye, M.R. Andrews, H.-J. Miesner, D.M. Stamper-Kurn, W. Ketterle, *Phys. Rev. Lett.* **82**, 2422 (1999)
17. J.L. Roberts, N.R. Claussen, S.L. Cornish, C.E. Wieman, *Phys. Rev. Lett.* **85**, 728 (2000)
18. J. Stenger, S. Inouye, D.M. Stamper-Kurn, H.-J. Miesner, A.P. Chikkatur, W. Ketterle, *Nature* **396**, 345 (1998)
19. M. Ueda, M. Koashi, *Phys. Rev. A* **65**, 063602 (2002)
20. H. Saito, M. Ueda, *Phys. Rev. A* **72**, 053628 (2005)
21. H. Saito, Y. Kawaguchi, M. Ueda, Private communication
22. D.M. Stamper-Kurn, M.R. Andrews, A.P. Chikkatur, S. Inouye, H.-J. Miesner, J. Stenger, W. Ketterle, *Phys. Rev. Lett.* **80**, 2027 (1998)
23. M.-O. Mewes, M.R. Andrews, D.M. Kurn, D.S. Durfee, C.G. Townsend, W. Ketterle, *Phys. Rev. Lett.* **78**, 582 (1997)
24. H. Schmaljohann, M. Erhard, J. Kronjäger, K. Sengstock, K. Bongs, *Appl. Phys. B* **79**, 1001 (2004)
25. C.J. Pethick, H. Smith, *Bose–Einstein Condensation in Dilute Gases* (Cambridge University Press, Cambridge, 2002)

⁴In our experiment, the upper bound of c_2 is estimated as $0.062a_B(4\pi\hbar/M)$ with spin-independent inelastic collisions assumed, where a_B is the Bohr radius. However, it is necessary for the estimation to take the spin-dependent inelastic collisions into consideration [21].